

186 66. (New) The method of claim 27, wherein step (a) contacting occurs in a pretreatment vessel and step (b) contacting occurs in a reaction vessel.

Please cancel claims 1-16 and 31 through 50 without prejudice.

### REMARKS

Applicants wish to thank Examiner Bekir Yildirim for the courtesy extended during a personal interview on January 31, 2003 at the U.S. Patent and Trademark Office, with the undersigned attorney for Applicants. Reconsideration of the above-identified application in view of the amendments above and the remarks following is respectfully requested.

Claims 17 and 27 have been amended; claims 1-16 and 31-50 have been canceled without prejudice; and claims 51-66 are added. Claims 17-30 and 51-66 are now pending.

Applicants amended claims 17 and 27 to add reference letters for each step of the process and/or for stylistic purposes. The amendment in no way changes the scope of claims 17 and 27 and was not made to address any issue of patentability of these claims.

Claims 50-66 have been amended to further define one or more embodiments of the invention that are more fully described in the specification on page 13, line 21 through page 14, line 8. Thus, these changes add no new matter. Applicants request entry of this amendment at this time.

For the reasons that follow, Applicants believe all claims are in condition for allowance and respectfully request reconsideration of this application.

#### I. Claims 17-30 and 50-66 Are Not Obvious In View of Kaiser.

The Examiner has rejected claims 17-30 in view of U.S. Patent No. 4,677,242 to Kaiser (Kaiser). Claim 17 is a method of making an olefin product from an oxygenate feedstock. It is a two step process. In one step, referred to as step a, a silicoaluminophosphate molecular sieve having a porous framework structure is contacted with a hydrocarbon at conditions effective to form at least an integrated hydrocarbon co-catalyst within the porous framework. In another step, referred to

as step b, the silicoaluminophosphate molecular sieve containing the integrated hydrocarbon co-catalyst is contacted with an oxygenate feedstock under conditions effective to convert the feedstock to the olefin product. The two step process is further characterized in that the silicoaluminophosphate molecular sieve has a catalytic activity index for methanol conversion at 250°C of at least 2.

Claim 27 is a method of making a polyolefin from an oxygenate feedstock. The method includes a two step process of producing an olefin product similar in scope to claim 17. Additionally, the olefin product is contacted with a polyolefin-forming catalyst under conditions effective to form the polyolefin. Claims 18-26, 51-56 and 63-64 depend from claim 17 and therefore contain all of the elements of claim 17. Claims 27-30, 57-62 and 65-66 depend from claim 27 and therefore contain all of the elements of claim 27.

Kaiser does not teach a step of contacting at conditions effective to form an integrated hydrocarbon co-catalyst as required by both claims 17 and 27. Kaiser teaches a step of contacting a silicoaluminophosphate molecular sieve catalyst in the presence of an aromatic diluent to produce an olefin product. The average kinetic diameter of the aromatic diluent is greater than the average pore size of the silicoaluminophosphate molecular sieve. Thus, the aromatic diluent cannot go into the pores of the catalyst. Kaiser does not have a separate step of forming an integrated hydrocarbon co-catalyst. Rather, Kaiser relies upon an external co-fed aromatic compound to increase desired product yield. It would not have been obvious in view of Kaiser to have a step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst.

Additionally, the present inventions set forth in claims 17 and 27 have an unexpected result over Kaiser. The initial step of forming at least an integrated hydrocarbon co-catalyst increases the amount of ethylene produced without letting the integrated hydrocarbon co-catalyst leave the catalyst into the product effluent stream. Thus, the product effluent stream has the benefit of an increased ethylene yield without increasing the amount of aromatic co-catalyst in the product effluent

stream. The Kaiser patent teaches the addition of aromatics, which pass through the pores of the catalyst into the product effluent stream. Regardless of the benefit, the product effluent stream in Kaiser has a more expensive clean up--particularly an aromatic recovery and recycle. The present method is particularly beneficial because it both increases the product yield and does not require an expensive aromatic recovery and recycle stream. Thus, the present invention has a benefit over the teaching of Kaiser.

Claims 17 and 27 as well as dependent claims 18-30 and 51-66 are patentable over Kaiser.

## II. Claims 17-30 and 50-66 Are Not Obvious In View of Brown et al.

The Examiner has rejected claims 17-30 in view of U.S. Patent No. 6,046,372 to Brown et al. (Brown). Brown does not teach a step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst as required by both claims 17 and 27. Brown teaches a step of contacting a feed, which contains methanol and/or dimethyl ether with a catalyst comprising a porous crystalline material in the presence of an aromatic compound under conversion conditions.

The porous crystalline material has a pore size greater than the critical diameter of the aromatic compound.

Brown does not have a separate step of forming an integrated hydrocarbon co-catalyst. Rather, Brown relies upon the aromatic compound during the conversion of methanol and/or dimethyl ether to a product containing C2 to C4 olefins that has higher product selectivity toward olefins. It would not have been obvious in view of Brown to have a separate step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst.

Additionally, the present inventions set forth in claims 17 and 27 have an unexpected result over Brown. The initial step of forming at least an integrated hydrocarbon co-catalyst increases the amount of ethylene produced without letting the integrated hydrocarbon co-catalyst leave the catalyst into the product effluent

stream. Thus, the product effluent stream has the benefit of an increased ethylene yield without increasing the amount of aromatic co-catalyst in the product effluent stream. Brown teaches the addition of aromatics, which pass through the pores of the catalyst into the product effluent stream. Regardless of the benefit, the product effluent stream in Brown has a more expensive clean-up--particularly an aromatic recovery and recycle. The present method is particularly beneficial because it both increases the product yield and does not require an expensive aromatic recovery and recycle stream. Thus, the present invention has a benefit over the teaching of Brown.

Claims 17 and 27 as well as dependent claims 18-30 and 51-66 are patentable over Brown.

### III. Claims 17-30 and 50-66 are not obvious In View of Kuechler I.

The Examiner has rejected claims 17-30 in view of U.S. Patent No. 6,137,022 to Kuechler et al. (Kuechler I). Kuechler I does not teach a step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst as required by both claims 17 and 27. Kuechler I teaches a separate step of contacting a porous crystalline material in the presence of one or more diluents that include aromatic compounds during the step of converting the oxygenates to olefins. Kuechler I does not have a step of forming an integrated hydrocarbon co-catalyst. It would not have been obvious in view of Kuechler I to have a step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst.

Additionally, the present inventions set forth in claims 17 and 27 have an unexpected result over Kuechler I. Kuechler I teaches no specific benefit of adding the aromatic as a diluent. As stated above, the present invention increases ethylene yield without adding aromatics to the product stream. Claims 17 and 27 both represent an important improvement over the teaching of Kuechler I. Claims 17 and 27 as well as dependent claims 18-30 and 51-66 are patentable over Kuechler I.

IV. Claims 17-30 and 50-66 are not obvious In View of Kuechler II.

The Examiner has rejected claims 17-30 in view of U.S. Patent No. 6,437,208 to Kuechler et al. (Kuechler II). Kuechler II does not teach a step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst as required by both claims 17 and 27. Kuechler II teaches a step of contacting a porous crystalline material in the presence of one or more inert diluents that include non-reactive aromatic compounds during the step of converting the oxygenates to olefins. Kuechler II further teaches a hydrocarbon co-feed that includes reactive alkyl aromatics as well as reactive aromatics. In Kuechler II, the diluent and co-feed occur during the step that oxygenates are converted to olefins and not in a separate step. Kuechler II, therefore, does not have a step of forming an integrated hydrocarbon co-catalyst. It would not have been obvious in view of Kuechler II to have a step of contacting at conditions effective to form at least an integrated hydrocarbon co-catalyst.

Additionally, the present inventions set forth in claims 17 and 27 have an unexpected result over Kuechler II and the other references. The Examples teach the benefit of practicing the present invention over the prior art. Different samples of a selected SAPO sieve were pretreated by exposing the SAPO sieve with different hydrocarbon compounds including acetone in vacuo at 200C for 1 hour. Results indicated that 1-3 wt.% hydrocarbon co-catalyst is contained within the pores. Example 2 describes the conversion of various catalyst samples including untreated SAPO sieve and SAPO sieve pretreated with acetone as described above. The data from this experiment is shown in Figure 5. The samples that were pretreated follow the process claimed in the present case.

Particularly, a silicoaluminophosphate molecular sieve having a porous framework structure is contacted with a hydrocarbon (i.e. acetone) at conditions effective (eg. 200C in vacuo for 1 hour) to form at least an integrated hydrocarbon co-catalyst within the porous framework. Next, the silicoaluminophosphate molecular sieve containing the integrated hydrocarbon co-catalyst is contacted with an oxygenate feedstock under conditions effective to convert the feedstock to the olefin product. The pretreated samples were compared against samples that

were not pretreated (i.e. as taught in accordance with prior art. The samples that follow the teaching of the present invention, by pretreatment with acetone, had an ethylene selectivity that was about 10 wt.% greater than the sample that was not pretreated at all reaction times shown.

Claims 17 and 27 as well as dependent claims 18-30 and 51-66 are patentable over Kuechler II.

Applicants invite the Examiner to telephone the undersigned attorney if there are any issues outstanding which have not been presented to the Examiner's satisfaction.

Respectfully submitted,

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Date



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APPENDIX A  
SPECIFICATION MARKED-UP TO SHOW CHANGES MADE

The paragraphs on page 4, line 31 to page 5, line 8 have been amended as follows:

FIG. 2 is a SSNMR spectrum of SAPO-34 catalyst containing alkylated single ring aromatics within the porous framework ~~is a plot comparing conversion of methanol by contacting methanol to SAPO catalysts with and without an integrated hydrocarbon co-catalyst;~~

FIG. 3 ~~is a plot of olefin selectivity for methanol reactions by contacting methanol to SAPO catalysts with and without an integrated hydrocarbon co-catalyst~~ is a SSNMR spectrum of SAPO-34 catalyst containing various hydrocarbons and alkylated single ring aromatics within the porous framework at various conditions;

FIG. 4 ~~is a SSNMR spectrum of SAPO-34 catalyst containing alkylated single ring aromatics within the porous framework~~ is a plot comparing conversion of methanol by contacting methanol to SAPO catalysts with and without an integrated hydrocarbon co-catalyst; and

FIG. 5 ~~s a SSNMR spectrum of SAPO-34 catalyst containing various hydrocarbons and alkylated single ring aromatics within the porous framework at various conditions~~ is a plot of olefin selectivity for methanol reactions by contacting methanol to SAPO catalysts with and without an integrated hydrocarbon co-catalyst.

The paragraph on page 12, line 1 to page 12, line 12 has been amended as follows:  
The hydrocarbon material that is contacted with or introduced into the silicoaluminophosphate molecular sieve must be capable of being converted to an integrated hydrocarbon co-catalyst which may be a single ring aromatic compound. The desired aromatic composition can be identified by Solid State Nuclear Magnetic Resonance (SSNMR) spectra comprising a peak in the 18-40

ppm region and a peak in the 120-150 ppm region. Preferably, the aromatic composition is selected from the ~~eroup~~ group consisting of alkyl substituted, single ring aromatics. Alternatively, if the intensity of the peak in the 18-40 ppm region is negligible, a single peak near 128 ppm also indicates a useful material of this invention, as this indicates the presence of benzene. Benzene also has the desired effect on catalytic activity, as it rapidly reacts with the oxygenate to make alkylated single ring aromatics.

The paragraph on page 13, line 3, tot page 13, line 20 has been amended as follows:

Hydrocarbon is introduced into the silicoaluminophosphate molecular sieve by contacting the silicoaluminophosphate molecular sieve or catalyst containing the silicoaluminophosphate molecular sieve with hydrocarbon in a pretreatment zone at conditions effective to form a composition comprising an integrated hydrocarbon co-catalyst which may be at least one single ring aromatic compound within the molecular pore structure of the silicoaluminophosphate molecular sieve. Within the pretreatment zone the conditions are such that an amount of an integrated hydrocarbon co-catalyst which may be a single ring aromatic will be formed within the silicoaluminophosphate molecular sieve pore structure that is effective in enhancing selectivity to ethylene or propylene in an oxygenate to olefin reaction process. The activity of a pretreated catalyst for oxygenate conversion can be expected to increase by a factor of at least about 20% relative to an untreated catalyst. The selectivity to ethylene or propylene of a pretreated catalyst in the oxygenate conversion reaction can be expected to increase by at least about 2 wt. %. Preferably, the an integrated hydrocarbon co-catalyst which may be a single ring aromatic will occupy at least 0.1 vol.% of the pore structure; more preferably, at least 10 vol.%; most preferably, at least 25 vol.%. the molecular pore structure—~~olefin will not be produced.~~



The paragraph on page 24, line 21 to page 24 line 24 has been amended as follows:

FIG. 5 shows the selectivity to  $C_2$ - $C_4$  olefins in the same experiment as described for ~~FIG. 5~~FIG. 4. The increase in prime olefin selectivity for methanol conversion for the catalyst of the invention, particularly the increase in ethylene selectivity, is shown to be about 10 wt.% or greater at any point in the experiment.

APPENDIX B  
CLAIMS MARKED-UP TO SHOW CHANGES MADE

17. A method of making an olefin product from an oxygenate feedstock, comprising:

(a) contacting a silicoaluminophosphate molecular sieve having a porous framework structure with a hydrocarbon at conditions effective to form at least an integrated hydrocarbon co-catalyst within the porous framework, and

(b) contacting the silicoaluminophosphate molecular sieve containing the integrated hydrocarbon co-catalyst with an oxygenate feedstock under conditions effective to convert the feedstock to the olefin product,

wherein the silicoaluminophosphate molecular sieve has a catalytic activity index for methanol conversion at 250°C of at least 2.

27. A method of making a polyolefin from an oxygenate feedstock, comprising:

(a) contacting a silicoaluminophosphate molecular sieve having a porous framework structure with a hydrocarbon at conditions effective to form at least an integrated hydrocarbon co-catalyst within the porous framework,

(b) contacting the silicoaluminophosphate molecular sieve containing the integrated hydrocarbon co-catalyst with an oxygenate feedstock under conditions effective to convert the feedstock to an olefin product, and

(c) contacting the olefin product with a polyolefin-forming catalyst under conditions effective to form the polyolefin,

wherein the silicoaluminophosphate molecular sieve containing the integrated hydrocarbon co-catalyst has a catalytic activity index for methanol conversion at 250°C of at least 2.